

Heats of Combustion of Liquid *n*-Hexadecane, 1-Hexadecene, *n*-Decylbenzene, *n*-Decylcyclohexane, *n*-Decylcyclopentane, and the Variation of Heat of Combustion With Chain Length¹

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The heats of combustion of five highly purified long-chain liquid hydrocarbons were measured with a bomb calorimeter. The calorimetric data yield the following values for the heats of combustion, $-\Delta H_c^\circ(25^\circ \text{C})$, of the liquid hydrocarbon with gaseous oxygen to form gaseous carbon dioxide and liquid water; 1 kilocalorie = 4.1840 kilojoule.

<i>n</i> -Decylbenzene	$= 2340.58 \pm 0.42$ kcal/mole
1-Hexadecene	$= 2519.17 \pm 0.44$
<i>n</i> -Hexadecane	$= 2557.15 \pm 0.42$
<i>n</i> -Decylcyclohexane	$= 2497.90 \pm 0.43$
<i>n</i> -Decylcyclopentane	$= 2347.54 \pm 0.46$

It is shown that, in the liquid as well as in the gaseous state, the increment in the heat of combustion (and hence in the heat of formation) per CH_2 group added in the *n*-alkyl side chain is a constant except for the first two members of the series, and that this increment has the same value for each of these series of compounds.

1. Introduction

This investigation is a continuation of the program [1, 2, 3, 4]² on the determination of some of the thermodynamic properties of compounds of importance in the national synthetic rubber program. In the study of the processes of polymerization and depolymerization, it is important to have thermodynamic data for the compounds in the liquid as well as in the gaseous state, and data on the variation of these properties with carbon chain length. Previous studies [5, 6] have shown that for several homologous series of gaseous hydrocarbons the heat of combustion increases by a constant amount upon the addition of each CH_2 group in the *n*-alkyl side chain; the first few members of each series are anomalous. In an earlier paper [5] the variation, with chain length, in the heat of combustion was studied for the liquid normal paraffins. The increment 156.26 kcal per CH_2 group was calculated, by the method of least squares, from these experimental data. This increment, together with previously reported values for the heats of combustion of the 1-alkenes [2, 22, 23, 24] alkylbenzenes [19, 20] *n*-alkylcyclopentanes [21], and *n*-alkylcyclohexanes [21] was used to calculate values for the heats of combustion of higher members of these series. Up to the present time there has been no experimental check on the validity of this extrapolation. Therefore, it seemed desirable to measure the heat of combustion of a higher member of each series. In this investigation the heats of combustion of 1-hexadecene, *n*-decylbenzene, *n*-decylcyclopentane, *n*-decylcyclohexane, and *n*-hexadecane were

measured. It was found that the measured heats agree, within the limits of experimental error, with those calculated using the increment 156.26 per CH_2 group added in the *n*-alkyl side chain.

This agreement shows that the heats of combustion of liquid hydrocarbons may be expressed by an equation of the form $\Delta H_c = A + Bm$, $m > 1$. A is a constant characteristic of the homologous series; B is the constant for the increase in the heat of combustion per CH_2 group added in the *n*-alkyl side chain, and is the same for all these series; and m is the number of carbon atoms in the side chain.

2. Units of Energy and Molecular Weights

The unit of energy upon which values reported in this paper are based is the absolute joule (j). For conversion to the conventional thermochemical calorie, the relation, 1 cal = 4.1840 j, is used.

The molecular weight of carbon dioxide, the mass of which was used to determine the amount of reaction, was taken as 44.010, from the 1952 table of international atomic weights [7].³

3. Method and Apparatus

The heats of combustion of the compounds were determined by burning the liquid samples in oxygen in a bomb, at constant volume. The procedure is similar to that used in previous investigations [9, 10,

¹ The work discussed herein was performed as a part of the research project sponsored by the Federal Facilities Corp., Office of Synthetic Rubber, in connection with the Government Synthetic Rubber Program.

² Figures in brackets indicate the literature references at the end of this paper.

³ The value for the atomic weight of carbon has recently been changed from 12.010 [7] to 12.011 [8]. The molecular weight of carbon dioxide was taken as 44.010 in this paper to retain consistency with values previously reported by the Thermochemical Laboratory for the heats of formation of other hydrocarbons and of carbon dioxide. The values of the heats of combustion per mole of hydrocarbon reported here can be corrected for the effect of the change to the new atomic weight of carbon by multiplication by the factor 44.011/44.010.

11,12]. The samples were sealed in weighed glass ampoules to prevent evaporation before the combustion period. The filled ampoules were weighed and placed in a platinum crucible in the bomb, 1 ml of water was added to the bomb, and the bomb was flushed and filled with purified oxygen to 30-atm pressure at 25° C. The internal volume of the bomb was 385 ml. The sample was ignited by means of an iron wire fuse, 0.012 cm in diameter and 5 cm long, coiled above the bulb. The amount of reaction was determined from the mass of carbon dioxide formed in the combustion. The heat of reaction was referred to the final calorimeter temperature of 28° C. Corrections were made for deviations from the standard calorimeter system, for the heat contributed by the ignition process of heating and burning the iron wire fuse, and for the heat produced by the formation of nitric acid during the combustion.

A series of calorimetric combustion experiments was made with each of the following substances: 1-hexadecene, *n*-decylbenzene, *n*-hexadecane, *n*-decylcyclohexane, *n*-decylcyclopentane, and benzoic acid (NBS Standard Sample 39g). All of the experiments were performed in as much the same manner as possible. The same calorimeter system, platinum resistance thermometer (NBS No. 485), and Mueller resistance bridge (NBS No. 24530) were used for all of the experiments. The experiments with standard benzoic acid were carried out to determine the energy equivalent of the calorimeter system. Some of these combustion experiments on benzoic acid were performed before and some after the experiments on the hydrocarbons, to allow for any errors which might result from changes in the calorimeter system with time.

The heat of combustion of *n*-hexadecane had been previously determined [10] in the Thermochemical Laboratory. The measurement was repeated as a general check on the procedure.

In several experiments, tests for completeness of combustion were made by analyzing samples of the product gases for carbon monoxide by the calorimetric method described by Shepherd [13].⁴ In no case was a significant amount of carbon monoxide found. A further check on the completeness of reaction was afforded by the ratio, *r*, of carbon dioxide produced in the combustion to the stoichiometric amount of carbon dioxide calculated from the mass of sample used. The following average values of *r* were obtained in the experiments; the number in parenthesis is the number of experiments for which the value of *r* was determined, and the uncertainty is the standard deviation of the mean:

1-hexadecene (5)	0.99982 ± 0.00011
<i>n</i> -decylbenzene (5)	0.99990 ± 0.00008
<i>n</i> -hexadecane (3)	1.00003 ± 0.00005
<i>n</i> -decylcyclohexane (4)	1.00019 ± 0.00012
<i>n</i> -decylcyclopentane (5)	1.00006 ± 0.00009
benzoic acid (5)	1.00008 ± 0.00003

⁴ These analyses were made by the Gas Chemistry Section of the Bureau.

4. Materials

The hydrocarbons were from the API-NBS series of purified hydrocarbons. The assigned purities of these compounds in mole percent, as determined from measurements of freezing points, were: 1-hexadecene, 99.84 ± 0.18; *n*-decylbenzene, 99.88 ± 0.10; *n*-hexadecane, 99.96 ± 0.04; *n*-decylcyclohexane, 99.86 ± 0.11; *n*-decylcyclopentane 99.80 ± 0.18.

The impurities in each compound are believed to be isomeric and present in such small amounts that their effect on the heat of combustion is less than the estimated uncertainty assigned.

5. Experimental Results

Table 1 lists the following data for the calibration experiments: The experiment number; the mass of benzoic acid; Δe_1 , the deviation from the standard calorimeter system; ΔR_c , the corrected increase in temperature of the calorimeter system; q_i , the correction for ignition energy; q_n , the correction for the formation of nitric acid; and E_s , the energy equivalent of the standard calorimeter system.

TABLE 1. Calibration experiments

Experiment No.	Mass of benzoic acid	Δe_1	ΔR_c	q_i	q_n	E_s
	<i>g</i>	<i>j/ohm</i>	<i>ohms</i>	<i>j</i>	<i>j</i>	<i>j/ohm</i>
1a	1.51226	15.1	0.286180	34.1	6.3	139799.7
2a	1.51221	16.5	.286084	34.3	5.7	139838.9
3a	1.51426	14.3	.286475	35.0	4.9	139839.1
4b	1.51471	5.5	.286751	36.1	21.0	139814.8
5b	1.51346	5.6	.286515	36.1	22.5	139819.9
6b	1.51315	5.6	.286432	35.9	22.0	139829.3
7b	1.51227	5.4	.286292	35.5	22.2	139815.8
8b	1.51279	5.5	.286390	35.0	22.0	139813.6
Mean.....						139821.4
Standard deviation of the mean.....						±4.8

^a Experiments performed before the combustion experiments on the hydrocarbons.

^b Experiments performed after the combustion experiments on the hydrocarbons.

The standard calorimeter system was considered to be the actual calorimeter as it existed just prior to the start of the first calibration experiment except that the sample to be burned was not considered a part of the system. Thus Δe_1 , the deviation from the standard calorimeter, was computed from the heat capacity of the benzoic acid, 1.21 j/g° C at 26.5° C (minus the heat capacity, 0.03 j/g° C ml, of that amount of oxygen at 30 atm and 28° C which was displaced by the benzoic acid) and the heat capacity of any parts of the calorimeter which were changed during the course of this investigation. Also included in Δe_1 are minor corrections for deviations of the average temperature of the experiment from the standard average temperature of 26.50° C (computed from the calculated change of heat capacity of the calorimeter with temperature) and for variations in the heat of the calibrating reaction due to deviations of the final temperature of the experiment from the standard final temperature of 28.00° C.

TABLE 2. Calorimetric combustion experiments

Experiment No.	Mass of carbon dioxide	Δe_2	ΔR_e	$Q(28^\circ \text{ C})$	q_i	q_n	$-\Delta U_B(28^\circ \text{ C})$	Mean value of $-\Delta U_B$ with its standard deviation
<i>n</i> -Decylbenzene								
	<i>g</i>	<i>j/ohm</i>	<i>Ohms</i>	<i>j</i>	<i>j</i>	<i>j</i>	<i>j/g CO₂</i>	<i>j/g CO₂</i>
1	2.85874	11.8	0.284165	39735.7	31.5	4.9	13887.0	13886.8 ±0.6
2	2.84816	10.4	.283114	39588.3	35.7	5.6	13885.1	
3	2.86925	10.6	.285230	39884.3	35.4	4.8	13886.6	
4	2.87300	10.4	.285654	39943.5	35.4	5.0	13889.0	
5	2.77388	10.2	.275735	38556.5	35.2	5.2	13885.3	
6	2.91076	10.9	.289374	40463.8	35.2	5.0	13887.6	
1-Hexadecene								
1	2.67126	10.9	0.285683	39947.7	35.8	6.0	14939.0	14941.1 ±0.6
2	2.72788	10.2	.291792	40801.7	35.9	6.0	14941.9	
3	2.66637	9.5	.285222	39882.8	36.2	5.7	14942.0	
4	2.65190	7.3	.283676	39666.0	35.7	5.9	14941.9	
5	2.60973	7.2	.279126	39029.8	36.4	5.7	14939.4	
6	2.68706	7.3	.287441	40192.5	35.8	6.0	14942.2	
<i>n</i> -Hexadecane								
1	2.73431	5.6	0.296839	41506.1	35.9	6.2	15164.3	15164.5 ±0.4
2	2.67456	6.8	.290381	40603.5	36.1	6.3	15165.5	
3	2.65107	6.1	.287786	40240.4	35.2	5.7	15163.5	
4	2.63905	5.1	.286517	40062.7	36.1	6.2	15164.7	
5	2.77207	5.0	.300919	42076.4	34.8	6.4	15163.8	
6	2.43637	5.0	.264537	36989.3	36.1	5.0	15165.3	
<i>n</i> -Decylcyclohexane								
1	2.71781	4.1	0.288249	40304.6	35.7	3.6	14815.3	14814.7 ±0.5
2	2.63256	3.7	.279230	39043.4	35.7	5.3	14815.4	
3	2.59134	3.6	.274887	38436.1	35.7	6.6	14816.2	
4	2.69554	6.0	.285858	39970.8	35.9	5.6	14813.1	
5	2.59597	3.5	.275319	38496.5	35.1	5.5	14813.7	
<i>n</i> -Decylcyclopentane								
1	2.48540	3.4	0.264257	36949.7	34.8	6.4	14850.1	14851.2 ±0.9
2	2.61842	3.5	.278461	38935.8	36.2	7.1	14853.4	
3	2.73536	1.9	.290933	40679.2	36.2	23.5	14848.3	
4	2.73981	1.8	.291485	40756.4	35.8	23.8	14853.9	
5	2.56356	0.5	.272715	38131.5	35.4	26.2	14850.4	
6	2.64616	.6	.281489	39358.4	35.5	24.6	14851.1	

The energy equivalent, E_s , was computed by:

$$E_s = [(Q'_B m_s + q_i + q_n) / \Delta R_c] - \Delta e_1,$$

where $Q'_B = \Delta U_B(28^\circ \text{C})$ is the heat of combustion of benzoic acid under the actual bomb conditions used in this investigation expressed in joules per gram of benzoic acid and m_s is the mass of benzoic acid. The value used for the heat of combustion of standard benzoic acid under standard conditions at 25°C is $Q_B(25^\circ \text{C}) = 26434.7 \pm 2.2 \text{ j/g}$.⁵

Under the actual bomb conditions and at 28°C , this value becomes $Q'_B(28^\circ \text{C}) = 26431.8 \pm 2.2 \text{ j/g}$.

Table 2 lists the following data for the calorimetric combustion experiments on the five compounds: The experiment number; the mass of carbon dioxide formed; Δe_2 , the deviation from the standard calorimeter system; ΔR_c ; $Q(28^\circ \text{C}) = (E_s + \Delta e_2) \Delta R_c$, the isothermal heat liberated by the process which took

place in the bomb; q_i ; q_n ; and $\Delta U_B(28^\circ \text{C}) = [Q(28^\circ \text{C}) - q_i - q_n] / \text{g CO}_2$, the heat of combustion in the bomb process expressed in joules per gram of carbon dioxide formed.

In table 3 the following quantities are given for each of the compounds: The mean value of $-\Delta U_B(28^\circ \text{C})$ expressed in joules per gram carbon dioxide from table 2, and in kilojoules per mole of hydrocarbon; $-\Delta U^\circ(28^\circ \text{C})$, the decrement in internal energy in the ideal bomb process [10]; $-\Delta H_c^\circ(28^\circ \text{C})$, the heat of combustion at constant pressure at 28°C ; $-\Delta H_c^\circ(25^\circ \text{C})$, the heat of combustion at constant pressure at 25°C ; and ΔH_f° , the heat of formation at constant pressure at 25°C . The uncertainties in the heats of combustion assigned to the values in table 3 were calculated using the relation,

$$\text{uncertainty} = [(0.0083)^2 + (2\sigma E_s)^2 + (2\sigma \Delta U_B)^2 + (0.01)^2]^{1/2}$$

where σ represents the standard deviation of the mean for the specified measurement, and all de-

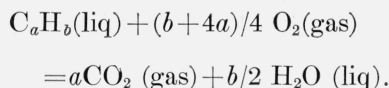
⁵ This value for benzoic acid, which is based on electrical calibration experiments made in the Thermochemical Laboratory [11], was used to maintain consistency with values previously reported [10] for the lower hydrocarbons, which were based on electrical calibrations. This value differs slightly from the value certified for benzoic acid, $Q_B(25^\circ \text{C}) = 26433.8 \pm 2.2 \text{ j/g}$ [14, 15]. However, the uncertainty thus introduced is within the estimated over-all uncertainty.

TABLE 3. *Heats of combustion and formation of the liquid hydrocarbons at 25° C*

Compound (liquid)	$-\Delta U_B$ at 28° C		$-\Delta U^\circ$ at 28° C	$-\Delta Hc^\circ$			ΔHf° at 25° C	
				at 28° C	at 25° C			
	<i>j/g CO₂</i>	<i>kJ/mole</i>			<i>kJ/mole</i>	<i>kJ/mole</i>	<i>kJ/mole</i>	<i>kcal/mole</i>
<i>n</i> -Decylbenzene.....	13886.8 ± 2.5	9778.5 ± 1.76	9775.1 ± 1.76	9791.3 ± 1.76	9793.0 ± 1.76	2340.58 ± 0.42	-219.12 ± 1.97	-52.37 ± 0.47
1-Hexadecene.....	14941.1 ± 2.6	10520.9 ± 1.85	10518.0 ± 1.85	10538.0 ± 1.85	10540.2 ± 1.85	2519.17 ± 0.44	-329.41 ± 2.09	-78.73 ± 0.50
<i>n</i> -Hexadecane.....	15164.5 ± 2.5	10678.2 ± 1.74	10675.5 ± 1.74	10696.8 ± 1.74	10699.1 ± 1.74	2557.15 ± 0.42	-456.35 ± 2.00	-109.07 ± 0.48
<i>n</i> -Decylcyclohexane.....	14814.7 ± 2.6	10431.9 ± 1.82	10429.0 ± 1.82	10449.0 ± 1.82	10451.2 ± 1.82	2497.90 ± 0.43	-418.39 ± 2.06	-100.00 ± 0.49
<i>n</i> -Decylcyclopentane.....	14851.2 ± 2.7	9804.0 ± 1.92	9801.3 ± 1.92	9820.1 ± 1.92	9822.1 ± 1.92	2347.54 ± 0.46	-368.19 ± 2.12	-88.00 ± 0.51

vations are expressed as percent of the measured quantity. The term 0.0083 percent is the assigned uncertainty in Q'_B ; the term 0.01 percent is arbitrarily assigned to allow for systematic errors in the measurements.

The heats of combustion refer to the reaction:



The heats of formation refer to the reaction:



The heats of formation were calculated from the heats of combustion using the values -68.317 ± 0.010 kcal/mole [16] for the heat of formation of liquid water and -94.052 ± 0.011 kcal/mole [14] for the heat of formation of carbon dioxide.

The value obtained for the heat of combustion of *n*-hexadecane (liquid), $-\Delta Hc^\circ(25^\circ \text{C}) = 2557.15 \pm 0.42$, is in good agreement with the value previously reported [10] by the Thermochemical Laboratory, $-\Delta Hc^\circ(25^\circ \text{C}) = 2557.58 \pm 0.68$ kcal/mole. The agreement of this value with those of other investigators has been described in a previous publication [5]. No previous measurements have been reported for 1-hexadecene, *n*-decylbenzene, *n*-decylcyclohexane, or *n*-decylcyclopentane.

6. Heats of Combustion of Homologous Compounds

The increment in the heat of combustion per CH_2 group added to the chain in the *n*-paraffins in the gaseous state has been shown [5] to have the constant value 157.44 ± 0.05 kcal/mole. This increment is constant beginning with the third member of the series, propane. From a study of the heats of isomerization of branched-chain paraffin hydrocarbons, it has been shown that the average bond energy of a particular bond between two atoms is affected, within experimental error, only by the other bonds (or atoms) attached to these atoms. Bonds (or atoms) further removed have no measurable effect on this bond energy unless major steric effects are present (such as are revealed by the difficulty in making a Fisher-Hirschfelder model of the molecule). The constancy in the increment per CH_2 group for members of the series above propane is in accord with this observation, since the addition of a CH_2 group to propane to form *n*-butane forms the same kind of bond and neighboring bonds as do further CH_2 additions. In later reports [6,18,19] the increment of 157.44 was applied to other homologous series of hydrocarbons with *n*-alkyl side chains to obtain heats of combustion for higher members of these series in the gaseous state.

A constant increment per CH_2 group for numbers above propane was also found [5] for the *n*-paraffins

TABLE 4.—*Comparison of experimental values of $-\Delta Hc^\circ$ with calculated values using the equation $-\Delta Hc^\circ = A + Bm$*

Series..... Structure.....	1-Alkenes Vinyl-(CH ₂) _m -H		Normal alkylbenzenes Phenyl-(CH ₂) _m -H		Normal alkylcyclopentanes Cyclopentyl-(CH ₂) _m -H		Normal alkylcyclohexanes Cyclohexyl-(CH ₂) _m -H	
	Experimental $-\Delta Hc^\circ$ (liq)	Deviation experimental -calculated	Experimental $-\Delta Hc^\circ$ (liq)	Deviation experimental -calculated	Experimental $-\Delta Hc^\circ$ (liq)	Deviation experimental -calculated	Experimental $-\Delta Hc^\circ$ (liq)	Deviation experimental -calculated
	<i>kcal/mole</i>	<i>kcal/mole</i>	<i>kcal/mole</i>	<i>kcal/mole</i>	<i>kcal/mole</i>	<i>kcal/mole</i>	<i>kcal/mole</i>	<i>kcal/mole</i>
<i>m</i> =0	-----	-----	780.98 ± 0.10	(+2.49)	786.54 ± 0.17	(+1.49)	936.88 ± 0.17	(+1.19)
<i>m</i> =1	-----	-----	934.50 ± 0.12	(-0.23)	941.14 ± 0.18	(-0.15)	1091.13 ± 0.23	(-0.80)
<i>m</i> =2	644.46 ± 0.18	+0.10	1091.03 ± 0.17	+0.06	1097.50 ± 0.22	-0.03	1248.23 ± 0.35	+0.06
<i>m</i> =3	800.61 ± 0.26	+0.01	1247.19 ± 0.16	-0.02	1253.74 ± 0.28	-0.03	1404.34 ± 0.27	-0.07
<i>m</i> =4	956.72 ± 0.33	-0.12	1403.46 ± 0.27	+0.01	-----	-----	1560.78 ± 0.29	+0.13
<i>m</i> =5	1112.87 ± 0.38	-0.21	-----	-----	-----	-----	-----	-----
<i>m</i> =10	-----	-----	2340.58 ± 0.42	-0.31	2347.54 ± 0.46	+0.09	2497.90 ± 0.43	-0.19
<i>m</i> =14	2519.17 ± 0.44	-0.07	-----	-----	-----	-----	-----	-----
<i>A</i>	-----	331.88	-----	778.49	-----	785.05	-----	935.69
<i>B</i>	-----	156.24	-----	156.24	-----	156.24	-----	156.24

*At saturation pressure.

in the liquid state. The value of this increment is 156.26 ± 0.05 kcal/mole. Because of possible variations in intermolecular forces, it was not evident that this increment could be applied to other homologous series of hydrocarbons with *n*-alkyl side chains in the liquid state. The values obtained here for the heats of combustion of some higher members of such series, together with values previously reported for the lower members in the liquid state, show that this value of 156.26 can be applied to these series as well. The constancy begins with the ethyl side chain.

The constant *B* was re-evaluated, by the method of least squares, utilizing all of the available experimental data from the Bureau. The new value of *B*, 156.24, is in agreement with the value 156.26 calculated from the *n*-paraffin series alone.

In table 4 are given the experimental values for the heats of combustion of compounds of several series in the liquid state. These data were obtained from the following sources: the heats of combustion of benzene, toluene, ethylbenzene, *n*-propylbenzene, and *n*-butylbenzene in the liquid state [19, 20]; the heats of combustion of cyclopentane, methylcyclopentane, ethylcyclopentane, *n*-propylcyclopentane, cyclohexane, methylcyclohexane, ethylcyclohexane, *n*-propylcyclohexane, and *n*-butylcyclohexane in the liquid state [21]; the heats of combustion of 1-butene [2] and 1-pentene [22] in the gaseous state together with heats of vaporization of 1-butene [23] and 1-pentene [24]; the heats of combustion of 1-hexene and 1-heptene in the liquid state [25]; and the heats of combustion of 1-hexadecene, *n*-decylbenzene, *n*-decylcyclohexane, *n*-decylcyclopentane, and *n*-hexadecane in the liquid state from the present investigation.

In table 4 are also given the deviations of these experimental data from the heats of combustion calculated using the equation $-\Delta H_c^\circ = A + Bm$ where *A* is the constant characteristic of the homologous series, *B* is the constant 156.24 kcal/mole for each series, and *m* is the number of carbon atoms in the *n*-alkyl side chain. The values of *A* were calculated weighting the value of ΔH_c° , for each compound measured, inversely as the square of its assigned uncertainty.

7. References

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